Coherent X-Ray Generation with Laser Driven Ions

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Abstract

Small parts of the electronic wavepacket of a multiple charged ion may still tunnel through the high Coulomb barrier for a sufficiently intense laser field of $10^{15}W/cm^2$ and higher. Solving numerically the corresponding Schrödinger equation we find that the periodic recollisions of such a wavepacket in the oscillating high power laser field will give rise to coherent X-ray radiation in the multiple keV regime, i.e. substantially higher than predicted or observed before.

Since the invention of the laser, scientists have tried to generate coherent light also far below the optical wavelength [1]. The striking advantages in high resolution measurements and imaging are doubtless. All X-ray lasers up to date however still lack the efficiency, size, price and in particular coherence properties of optical laser schemes. In one of the most promising mechanisms for coherent high frequency generation, high power near optical laser fields were imposed on atomic systems. Parts of the electron wavepackets are then oscillating through the ionic core and due to the nonlinear interaction with the nucleus give rise to multiples of the applied laser frequency, i.e. harmonics [2]. In the so called tunneling regime only very small parts of the wavepackets tunnel through the laser modified Coulomb barrier but when returning the so far highest orders of harmonics were observed [3,4]. The highest possible achievable photon energy has been determined theoretically to give the so called cut-off law [5–7]

$$n_{max}\hbar\omega = I_p + 3.2U_p \tag{1}$$

with the ionisation potential I_p, the ponderomotive energy $U_p = E^2/4\omega^2$, $\hbar$ the Planck constant, n_{max} the maximal order of harmonics and E and ω being the maximal amplitude and angular frequency of the applied laser field.

The above simple formular was then explained in a simple three step model [8]: (i) parts from the bound electron wavepacket tunnel through the Coulomb barrier at the time when the laser field is close to its maximum and is sufficiently narrow for a substantial tunnel rate (ii) the born wavepackets oscillates freely and classically in the laser field and when returning has picked up the kinetic or ponderomotive energy $3.2\,U_p$ and (iii) at the return with the nucleus may at best drop back to the ground state of energy $-I_p$ and release in form of radiation the ionisation energy I_p and all the gained kinetic energy $3.2\,U_p$. If this process is repeated several times, interference among those processes leads to coherence of the generated light [9].

Many attempts have been made to enhance the above cut-off rule for the benefit of even higher coherent high frequency generation. By preparing the atomic ensemble appropriately

prior the interaction with the laser field or turning to more complex systems as molecules, the factor 3.2 could be enhanced to at best the order of 10 [9]. In principle the U_p could be strongly increased by several orders of magnitude with present laser technology by increasing the driving laser field strength up to the relativistic regime [10]. However, the Coulomb barrier of neutral atoms is then modified so strongly that the above described three step model does not apply anymore and instead often only Bremsstrahlung harmonics arise.

A large experimental enhancement was recently achieved independently by the groups of Krausz [11] and by that of Murnane and Kapteyn [12] where ultra short pulses were employed. With at about 500 eV they have generated the shorted coherent wavelength so far and have entered the so called water window where optical imaging of living species in water is possible. The break through was possible essentially because of the use of very short pulses which allowed to move quickly to the highest intensity of the laser pulse before too much of the ground state wavepacket was lost. In spite of this dramatic improvement a draw back of the short pulses is that only few recollisions of born wavepackets with the atomic core are possible and thus the coherence properties of the generated light are limited.

In this Letter we present a way to enhance high harmonic generation towards the hard X-ray regime, maintaining still high coherence properties. For this purpose we propose to use multiply charged ions [13,14] rather than atoms because they possess a considerably larger ionization potential I_p and according to the cut-off rule Eq.(1) promise a far larger maximal harmonic order. Since relativistic corrections turned out negligible for the low ion charges of interest here, we solve numerically the corresponding Schrödinger equation. For each effective ion charge we evaluate the appropriate laser electric field strength to allow for tunneling and recollision harmonic generation. For a charge of $Z = 3$ of the ionic core as sensed by the active electron, harmonics up to the order 2500 arise, i.e. with a photon energy of more than 4 keV.

The parameter that has been employed so far to estimate if the laser and atomic parameters are such that the three step tunnel mechanism takes place is the Keldysh parameter γ_K [9]

$$\gamma_K = \sqrt{\frac{Z^2 I_p^{(1)}}{2U_p}} = \frac{Z\omega}{E}\sqrt{2I_p^{(1)}} = \frac{Z\omega}{E} \tag{2}$$

with $I_p = Z^2 I_p^{(1)}$ being the ionization potential of the ion of interest with effective charge Z and $I_p^{(1)}$ the ionization potential for hydrogen. If this parameter is well below unity, one is in the tunneling regime. To have significant tunneling and thus harmonic intensity, we need a small γ. However to avoid substantial ionization this parameter should not be too small. Following ref. [16] tunneling decreases exponentially with Z^3 and a U_p increasing with Z^6 is necessary to compensate for this. As this is only an estimation for our model potential we have considered and optimized the tunneling dynamics also via numerical studies prior calculating the spectrum. We show in fact that we have to decrease the γ parameter with increasing charge of the ion strongly to ensure a sufficiently large tunneling rate and thus high harmonic output.

The laser field strength employed do not exceed the order of $2 \cdot 10^{16} W/cm^2$ so that due the laser field we do not enter the relativistic regime. Concerning the interaction of the electronic wavepacket with the ionic core we employ a net charge of up to $Z = 3$. This can also be assumed as non relativistic; a corresponding calculation via the Dirac equation will thus give essentially equal results. For more highly charged ions the velocities close to the nucleus can be significant as compared to the speed of light c so that the laser Lorentz force term can be nonnegligible as compared to the laser electric field [15]. This term would then give rise to complex modifications to the recollision dynamics and thus to the harmonic spectrum but is not of interest to the nonrelativistic regime of interest here [17].

With those assumptions in mind we consider the following dynamical equation for the electronic wavefunction $\Psi(x,t)$ as a function of the spatial coordinate x in the direction of the linearly polarized laser light and the time t:

$$i\partial_t \Psi(x,t) = H_S \Psi(x,t) \tag{3}$$

where H_S stands for the Schrödinger Hamiltonian with:

$$H_S = \frac{1}{2}\left(p - \frac{A(t)}{c}\right)^2 + V(x,Z) \tag{4}$$

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where the vector potential of the laser pulse is defined via $A(t) = E \cdot h(t) \cdot \sin(\omega t)$. The electric field $E \cdot h(t)$ involves a pulse shape $h(t)$ with turn-on phase and a phase with constant amplitude during which we evaluate the radiation spectrum.

We use the regularized nuclear potential,

$$V(x, Z) = -\frac{k}{\sqrt{x^2 + s}} \tag{5}$$

where $s = 2$ is a smoothing parameter of the nucleus to compensate for the neglect of the second and third dimension [18]. k is a direct function of the net charge Z of the ion and can be adjusted to recover the experimental values for the binding energies.

In our numerical analysis for hydrogen (H) and hydrogen like ions (He$^+$, Li^{2+}) interacting with short intense laser pulses, we employed a laser wave length of $\lambda = 800$nm (TiSapphire), an electric field amplitude with intensity $I \sim 10^{14} - 10^{16}$W/cm^2 and pulse lengths between 20 and 200 cycles with a turn-on phase of 5 to 20 cycles. In fig. 1 we displayed the effective potentials for H, He$^+$ and Li^{2+} in the laser pulse at the time of maximal electric field and as a horizontal line the corresponding ground state energy. For the case of hydrogen we have chosen the laser parameters ($\gamma_K = 0.89$) such that we are well in the tunneling regime but simultaneously such that ionization is still small. When moving to He^+ we first increased the electric field strength by a factor of two only to obtain the same Keldysh parameter and note from the corresponding dashed line for the effective potential in Fig. 1 that the tunneling barrier is then very large and that we appear to be in the intermediate regime to the multiphoton regime. While the ionization potential scales with Z^2, the tunneling rate reduces expotenially with Z^3/E [16]. In the solid line for He^+ we have further increased the electric field strength such that we have similar ionization as for H above, however though with a smaller γ_K.

Before addressing Li^{2+}, we discuss Fig. 2 where we have displayed the radiation spectrum corresponding to the situations in H (a) and the two cases for He^+ displayed in Fig. 1. For H we are in the tunneling regime and thus find as well known a plateau and cut-off following Eq. (1). In Fig 2(b) for increasing I_p and U_p by a factor of two we find a

spectrum with the expected cut-off enhanced by a factor of $Z^2 = 4$. However, even though γ_K is unchanged we have clearly moved towards the multi photon regime. The harmonic structure has detoriated and we find even resonance structures in the low frequency part of the spectrum. The enhancement of about the 12th harmonic corresponds to the transition from the first excited to the ground state. Also we note a beginning of a tilting of the spectrum in the plateau area. To reenter the tunneling regime, we have increased the E field further, such that the ionization rate becomes comparable to that for H in Fig. 1 a) and found an up-conversion of the cut-off frequency by a factor of 10.

We continue the procedure for Li^{2+} as depicted in the lowest entry of Fig. 1. We have increased the electric field with respect to H not only by a factor 3 but substantially more to secure sufficient tunneling. However, for such high harmonics as expected here and the corresponding low efficiency we chose a long pulse for a clear resolution of the very high harmonics. This requires us to maintain a sufficiently wide ionization barrier somewhat larger than in the two previous cases and thus accept a spectrum with a less pronounced cut-off frequency. Still we achieve at about 2500 harmonics as visible in Fig. 3, i.e. hard X-ray harmonics with an energy of at about 4 keV. We note that sometimes in the literature those energies are still counted to the upper end of soft X-rays. However, proceeding to higher charged ions, even higher harmonics are achievable, though with an increasingly small efficiency. Given the available parameters we note a scaling law of at about Z^3 for the energy of the cut-off harmonics as a function of the ion charge and the correspondingly chosen laser field intensity. We note that for practical reasons we have increased the intensity with rising charge Z rather than modifying the frequency which is less controllable experimentally. A constant wavelength of the applied laser field may however be problematic, since relative to the modified characteristic length of the ionic core due to the variable Z, one may need adapt the time available for tunneling.

Moving towards the relativistic regime [10] with higher laser intensities and ionic charges, higher harmonics are possible. However, the harmonic spectrum deviates further from the traditional structure in the tunneling regime including a horizontal plateau and a well pro-

nounced cut-off. The magnetic component of the laser field induces a significant momentum transfer in the propagation direction of the laser field. This makes recollisions more difficult, especially involving long return times after many free oscillations in the laser field. As a consequence we find a reduction of harmonics between the perturbative and the cut-off regime. This goes beyond the scope of this letter and will be discussed in detail in future work [17].

In conclusion multiply charged ions still allow for tunneling and recollisions of electron wave packets with the parent ionic core with laser field intensities well above those employed before for high harmonic generation with atoms. Consequently coherent X-rays become feasible in the multiple keV regime. The efficiency is below that for coherent high harmonic generation via laser driven atoms.

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Figure Captions

Fig.1. The effective potentials for $Z = 1$ (H, k=1), $Z = 2$ (He$^+$, k=3.48) and $Z = 3$ (Li^{2+}, k=7.35) interacting with electric fields corresponding to the intensities $1.4 \cdot 10^{14}$W/cm^2 ($\rightarrow \gamma_K = 0.89$ for H), $2.19 \cdot 10^{15}$W/cm^2 ($\rightarrow \gamma_K = 0.456$ for He$^+$) and $2 \cdot 10^{16}$W/cm^2 ($\rightarrow \gamma_K = 0.255$ for Li^{2+}). The corresponding ground state energies are indicated by the horizontal lines. The laser wavelength is 800nm, which corresponds to an atomic frequency $\omega =$0.057au. He$^+$ with the same γ_K as H is represented by the dashed line.

Fig.2. The harmonic spectrum for H (a) and He$^+$ (b/c) in appropriately intense laser fields as given in Fig. 1. The pulses consist of 20 cycles full intensity and 5 cycles turn-on. With Eq. 1 the cut-off is placed at $n_{max,H} = 27$ for H, $n_{max,He^+} = Z^2 n_{max,H} = 108$ for He$^+$ with the same γ_K as for H and $n_{max,He^+} = 305$ with the enhanced U_p (the ionisation rate here is comparable to that of H and is about 20%).

Fig.3. a) The harmonic spectrum for Li^{2+} for the laser parameters given in Fig. 1. b) is an enlargement of the high frequenct range of figure a). The full intensity was applied here for 150 cycles with an 18 cycle turn-on. After 100 cycles we have $\sim 60\%$ ionised and just after 150 cycles essentially 100% is ionised.
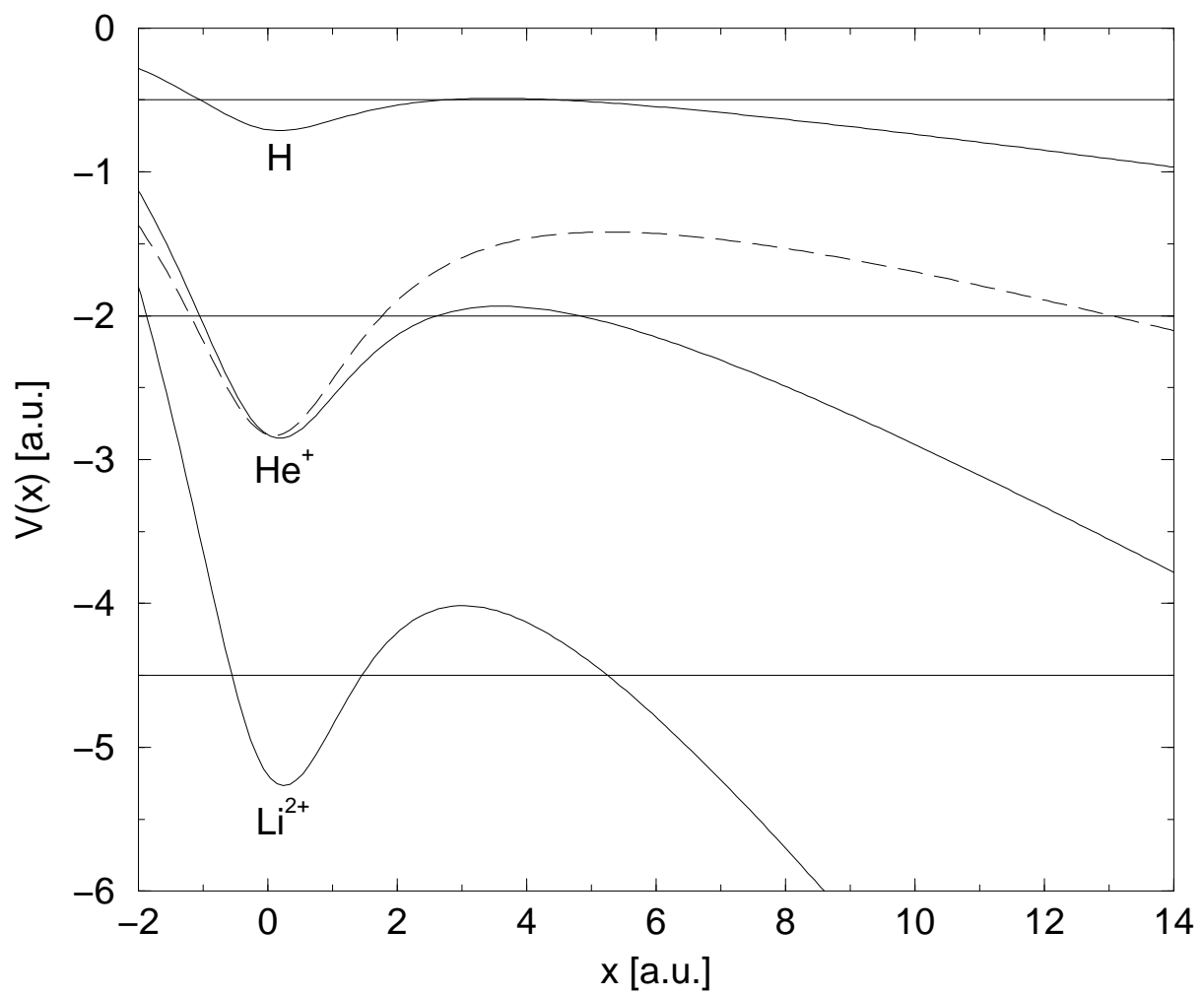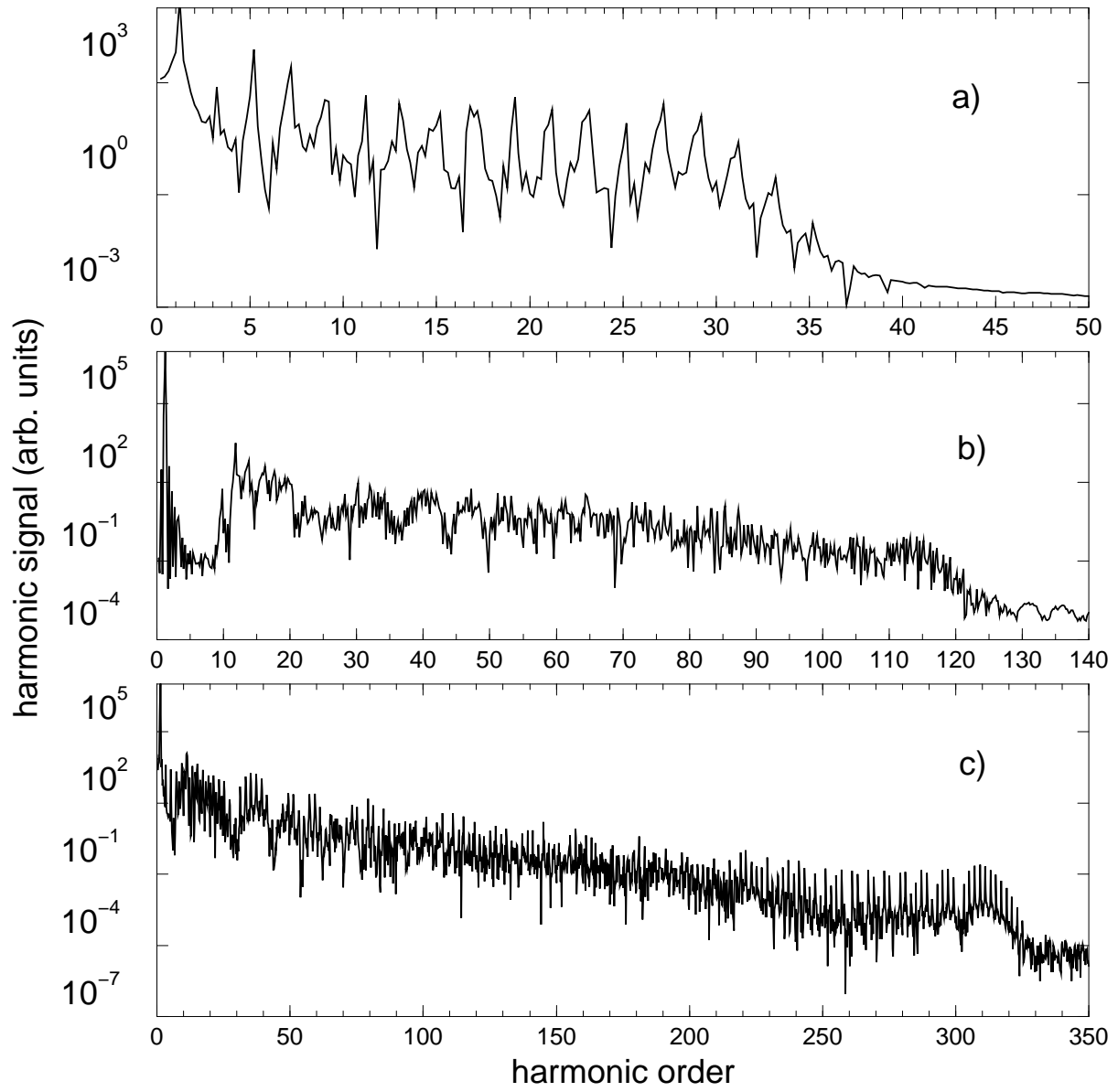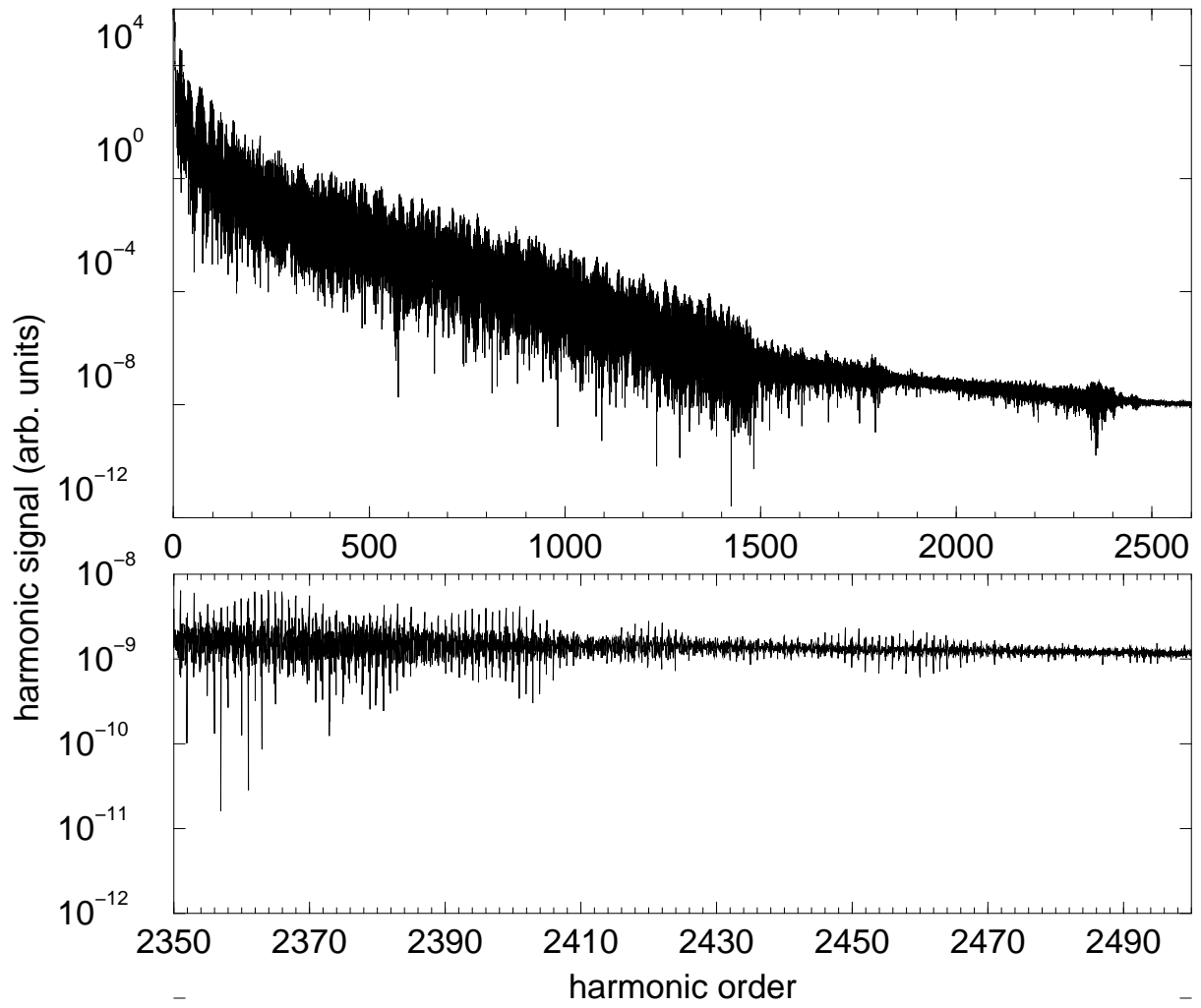
FIG. 1. M. Casu et al., "Coherent ..."

FIG. 2. M. Casu et al., "Coherent .."

FIG. 3. M. Casu et al., "Coherent .."